Breakdown of LS Coupling for a Parity Unfavored Transition in Neon: Angle Resolved 2D Imaging of Two Electrons Processes

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INTRODUCTION

The mechanism by which two electrons can be simultaneously excited by a single photon has been subjected to significant experimental and theoretical investigations over many years. Since this process only occurs via inter-electron correlations, the many body dynamics of an atom can be probed. These studies have, for the most part, focused on helium [1], as it represents the simplest system for which two-electron processes can be investigated. Systems in which the ground state electrons occupy higher angular momentum orbitals provide the opportunity to observe more complex effects since there are many more optically allowed transitions. Until the recent availability of third generation sources, the study of doubly excited states in heavier atoms was limited by the achievable resolving power [2,4]. The first photoabsorption measurements in neon [5] indicated that all of the structure due to two electrons excitation, with only one exception, could be well described by LS-coupling. However, a recent very high resolution photoionization experiment, below 53 eV [6], exhibited rich resonance structure which could not be explained in a simple LS coupling picture. This corroborates the general importance of LS coupling breakdown in neon pointed out by Samson et al. [3]. In the present study, we have concentrated on achieving a complete picture of the excitation and decay processes of doubly excited states in neon,

$$h\nu + Ne\ (2s^2 2p^6\ {}^1S_0) \rightarrow Ne^{**}\ (2s^2 2p^4\ nln'l')$$
$$\rightarrow Ne^{+*}\ (2s^2 2p^4\ n''l'') + e^-$$

by measuring the ejected electrons' energies as a function of exciting photon energy while simultaneously determining their angular distributions.

EXPERIMENTAL TECHNIQUE

We used a technique that combines for the first time, angle-resolved time-of-flight (TOF) spectrometry with two-dimensional (2D) imaging. Specifically, this method uses two highly efficient TOF energy analyzers [7] to produce angle-resolved, two-dimensional images of electron emission processes. The TOF spectrometers are set at 0° and 54° with respect to the electric field vector of the incident radiation from the High Resolution Atomic, Molecular and Optical (HRAMO) undulator beamline (9.0.1) at the Advanced Light Source at Lawrence Berkeley National Laboratory.

RESULTS AND INTERPRETATION

A part of the overall data collected is shown in Fig. 1. It shows the population of three ionic states, (a) $2s^2p^6$ (^{2}S), (b) $2s^22p^4$ (^{3}P), and (c) $2s^22p^4$ (^{3}P)3s(^{2}P), appearing along lines of constant binding energy parallel to the photon energy axis. Resonant enhancement is clearly seen at a number of well defined photon energies, as indicated by localized changes in the shade levels. More conventional one dimensional spectra showing the population of a given ionic state as a function of photon energy can easily be extracted from the data of Fig. 1 for more quantitative analysis.

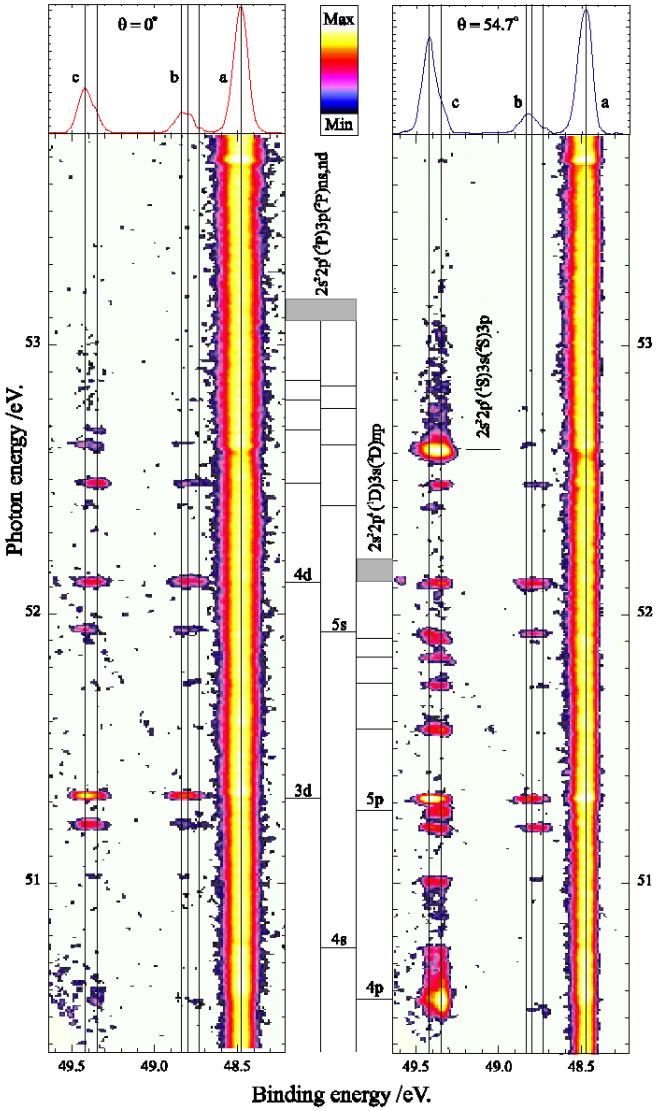

Figure 1. 2D images of electron emission processes taken at 0° and 54.7°. The images represent the electron yield as functions of the electron binding energy and the photon energy. The positions of three doubly excited Rydberg series are indicated in between the two images.

Such spectra are presented in Fig. 2 along with the derived asymmetry parameter β values. In this figure, three Rydberg series are observed which clearly have different behavior at the two different angles giving rise to mostly negative β values. The collected data, when viewed with the 2D representations at both angles (as seen in Fig. 1), allows the easy identification of certain unexpected features. For example, the angular distribution of electrons corresponding to the $2s^2 2p^4(^3P)3s(^2P)$ continuum is such that it is absent in the Θ = 0° spectrum throughout the range 50.5–60 eV, with the general exception of the decay of members of two Rydberg series, $2s^2 2p^4(^3P)3p(^2P)$ ns,nd as shown in Fig. 1 and Fig. 2. From angular momentum transfer consideration [8] this ionic state is produced by a parity-unfavored transition with only one outgoing wave (εp) in LS-coupling. These class of transitions are characterized by angular distributions in which the electrons are predominantly ejected in a direction orthogonal to the electric field, with angular asymmetry parameter β=-1. This means that the appearance of any signal at Θ=0° as can be seen from Fig. 1 and 2, is an immediate indication of the breakdown of LS coupling for these resonances. Preliminary R-matrix calculations for this process had difficulty

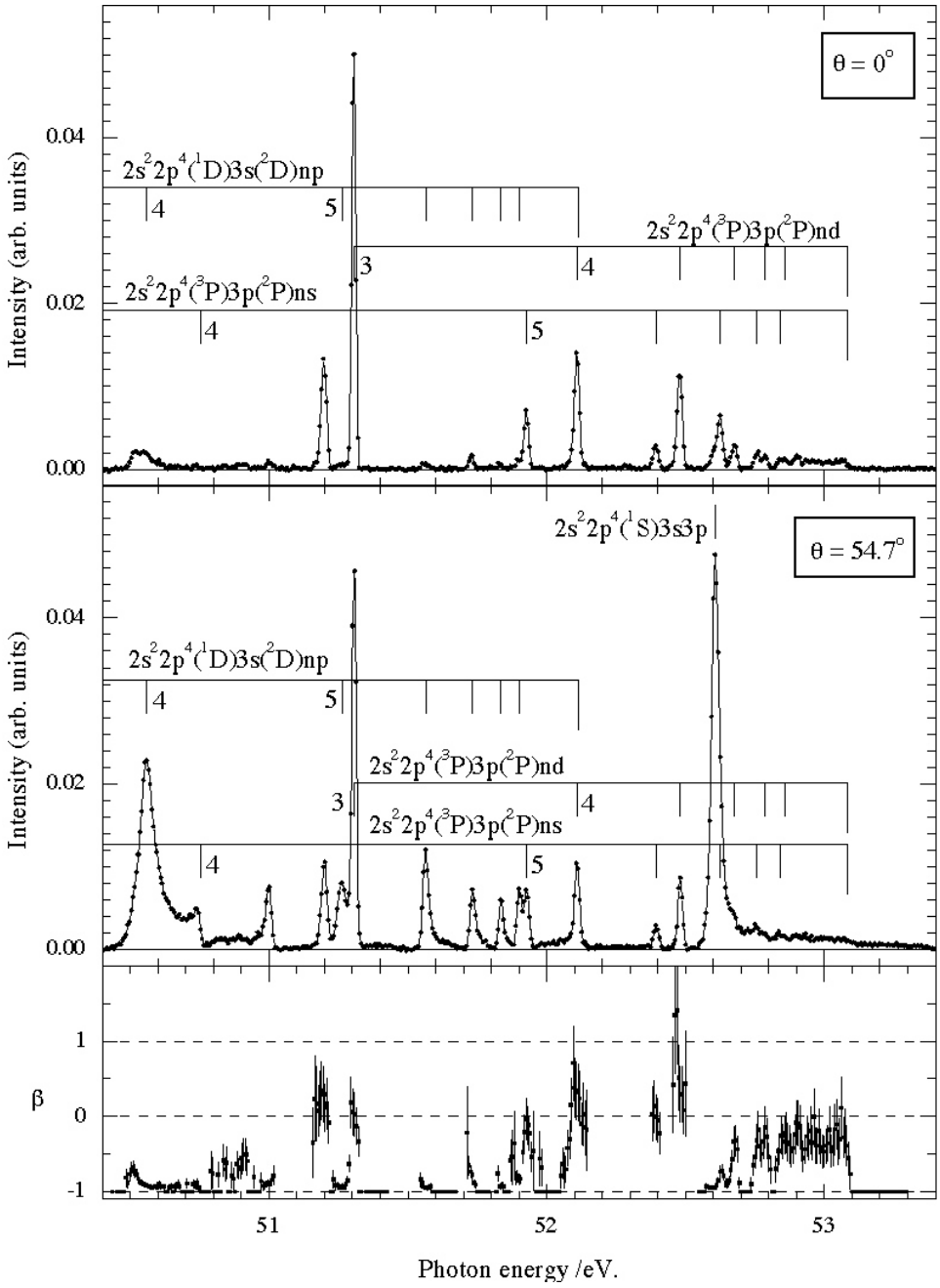

Figure 2. Spectra showing the $2s^2 2p^4(^3P)3s(^2P)$ ionic state as a function of photon energy at 0° and 54.7° along with the derived β values.

giving accurate predictions for the above deviation from LS coupling since extensive configuration interaction and spin-orbit effects are both required. We have also measured (using data from Fig. 1) a non-constant ratio of fine structure levels, unexpected in such a light atom, but observed previously in higher-Z [9] atoms since spin-orbit effects are expected to play a significant role.

Important implications of our findings [10] are twofold. First, viewing 2D photoionization images at various angles permits the easy identification of any unexpected behavior, such as the LS-forbidden, that might be overlooked using less-detailed probing techniques. Second, detection of prominent spin-orbit effects means that it is not safe in general to assume the validity of LS

coupling, even for a system as light as neon, when performing detailed theoretical calculations, spectroscopic assignments, or experimental calibrations.

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